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TITLE: Airplane and Drop Experiments on Crystallization of  $\text{In}_x\text{Ga}_{1-x}\text{Sb}$  Semiconductor under Different Gravity Conditions

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## Airplane and Drop Experiments on Crystallization of $\text{In}_x\text{Ga}_{1-x}\text{Sb}$ Semiconductor under Different Gravity Conditions

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### ABSTRACT

Melting and crystallization experiments of InGaSb were done under the reduced gravity condition ( $10^{-2}\text{G}$ ) in an airplane and at the normal gravity condition (1G) in the laboratory. Crystallized InGaSb was found to contain many needle crystals in both the cases. Reduced gravity condition was found to be more conducive for crystal growth than the normal gravity condition. Formation of spherical projections on the surface of InGaSb during its crystallization was in-situ observed using a high speed CCD camera in the drop experiment. Spherical projections showed dependence of gravity during its growth. Indium compositions in the spherical projections were found to vary depending on the temperature.

### INTRODUCTION

$\text{In}_x\text{Ga}_{1-x}\text{Sb}$  is a potential optoelectronic device oriented material and it could be used to fabricate commercially viable detectors, thermo-photo-voltaic (TPV) cells.  $\text{In}_x\text{Ga}_{1-x}\text{Sb}$  along with its binary counterparts GaSb and InSb are interesting III-V model materials for space ventures because of their low melting temperatures ( $712^\circ\text{C}$  and below) and low vapour pressures. It is extremely difficult to grow high quality  $\text{In}_x\text{Ga}_{1-x}\text{Sb}$  bulk crystals on earth due to gravity induced effects. As the densities of the components are different, solute transport occurs due to buoyancy. For this reason, microgravity condition in space is ideally suited to grow high quality and defect free  $\text{In}_x\text{Ga}_{1-x}\text{Sb}$  crystals as the gravity induced negative effects can be overcome [1-3]. In order to investigate the effect of gravity on the dissolution and crystallization processes, we carried out two microgravity experiments. The first one was performed in the Second International Microgravity Laboratory (IML-2) in 1994 [4-7]. We studied the effects of diffusion and convection on the melt mixing of In/GaSb/Sb. One of the important observations made in this venture was the formation of many circular projections on the surface of the InGaSb spherical sample. The indium compositions in this area were different from those in the main body of the sample. The other experiment was carried out in a Chinese recoverable satellite in 1996 [8-10].

It is not always possible to venture microgravity experiments in space using space shuttles due to economic and other major constraints. Hence, it is necessary to find alternate avenues to perform this type of experiments on earth itself using the facilities like drop tower, airplane etc. It is also important to know how the crystal growth processes take place under the reduced gravity conditions like  $10^{-1}\text{G}$ ,  $10^{-2}\text{G}$

etc. and under the higher gravity conditions, like, 2G, 3G etc. In the present article, we discuss about the results of the airplane and drop experiments on the crystallization of InGaSb conducted under different gravity conditions.

## EXPERIMENTAL

For the airplane experiment, polycrystalline  $\text{In}_x\text{Ga}_{1-x}\text{Sb}$  ( $x=0.05$ ) cut into a dimension of  $5 \times 5 \times 0.2 \text{ mm}^3$  was used as the starting material. The sample was sandwiched between two thin quartz glass plates and was sealed using high temperature adhesives. Thermocouples were connected to the samples to monitor and control the temperature during the melting and crystallization processes. The sample configuration is shown in Fig.1. An equipment was designed and constructed for the airplane experiment. The sample was mounted on a movable platform so that the position of it could be changed using a motor during the experiment. The InGaSb sample was heated to a temperature of  $706^\circ\text{C}$ . During this time, InGaSb poly crystal melted and changed to In-Ga-Sb solution. When the gravity level changed to  $10^{-2}\text{G}$ , the sample was moved out of the furnace and hence the temperature decreased rapidly. This process is schematically represented in Fig.2. During the rapid decrease of temperature, InGaSb crystallized. The complete experiment was videographed using a high resolution CCD camera. The same type of experiment was carried out under the normal gravity (1G) in the laboratory.

For the drop experiment, polycrystals of InGaSb were used as the starting materials. The sample structure was similar to Fig.1. This experiment was performed using a 150m drop tower facility present in the Micro-Gravity Laboratory of Japan (MGLAB). An equipment comprising a furnace to heat the sample, thermocouples, temperature controllers to control the sample and furnace temperatures, a high speed and high resolution CCD camera, and two lights, was constructed for this study and the schematic diagram of it is shown in Fig.3. The melting and crystallization of InGaSb during the experiment was videographed using the high speed and high resolution camera. The recorded images and the corresponding temperatures and gravity levels were used to study the melting and crystallization processes. InGaSb polycrystalline plate sample was heated to a temperature of  $800^\circ\text{C}$  at a rate of  $50^\circ\text{C}/\text{min.}$  and kept for 10 minutes. Later, the sample was cooled down by cutting off the power supply. During the cooling period, molten InGaSb started crystallizing. During the crystallization, the capsule was dropped in to the vacuum tower.

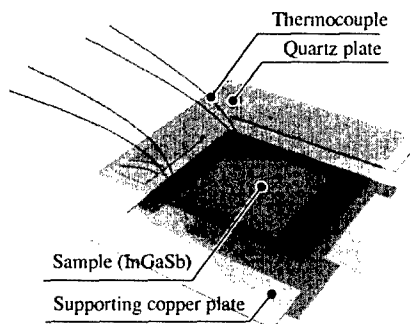


Fig.1. Sample configuration for the airplane and drop experiments.

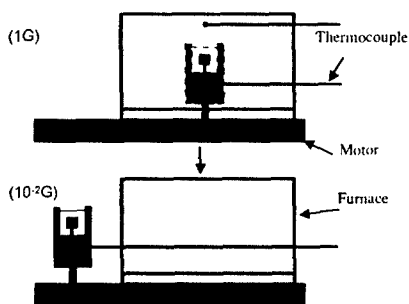


Fig.2. Change of sample position during the airplane experiment.

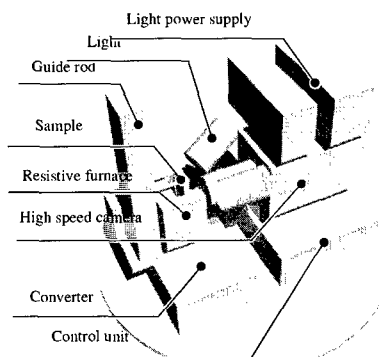


Fig.3. Schematic diagram of home-made equipment for the drop experiment.

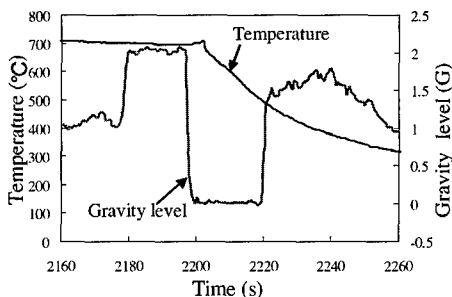


Fig.4. Variation of gravity level and sample temperature with time in the airplane experiment.

## RESULTS AND DISCUSSION

### Effect of gravity on the needle crystal growth

Figure 4 shows the variation of the gravity level with the flying time of the aircraft and the temperature of the InGaSb sample during the experiment. The set temperature of the sample was decreased to less than  $500^{\circ}\text{C}$  in a time of a second after the gravity became  $10^{-2}\text{G}$ . Figure 5 shows the photographs of the crystallized InGaSb samples under  $10^{-2}\text{G}$  and  $1\text{G}$  conditions, respectively, and the corresponding EPMA line profiles of indium composition measured on the surfaces of the samples. The needle crystals were found to have formed during the cooling of InGaSb. This result is similar to the needle crystal formed in our space experiment under microgravity condition [10]. In the case of the needle crystal formed under reduced gravity ( $10^{-2}\text{G}$ ) and normal gravity, indium composition should be as small as 0.005. Figure 6 shows the binary phase diagram of the InSb-GaSb system. As marked in the phase diagram, the sample must be cooled down fast to get the crystal of indium composition 0.005. Hence, in the EPMA profile, the portions where the In composition is 0.005, correspond to the InGaSb needle crystal and the portions where the In composition is high, correspond to the residual solution. The needle crystal is surrounded by the residual solution and its indium composition is much more than that of the crystal. As seen in the EPMA profiles, in the case of reduced gravity processed sample, the number of indium peaks is relatively smaller than the peaks observed in the normal gravity processed sample.

Figures 7(a) and (b) show the statistics of the size of the needle crystals formed under reduced gravity and normal gravity conditions, respectively. As clearly seen in the figures, the number of smaller sized crystals formed under normal gravity conditions is much more than the number of smaller crystals formed under reduced gravity condition. It is obvious that the convective forces during the crystal growth processes behave differently depending on the gravity conditions. Under the reduced gravity condition, growth of larger sized crystals is more feasible as the negative effect of the convective forces is relatively less. In the case of the crystal growth under normal gravity, due to the convective forces, most of the needle crystals grown were smaller. This clearly indicates that the reduced gravity condition is better suited for crystal growth.

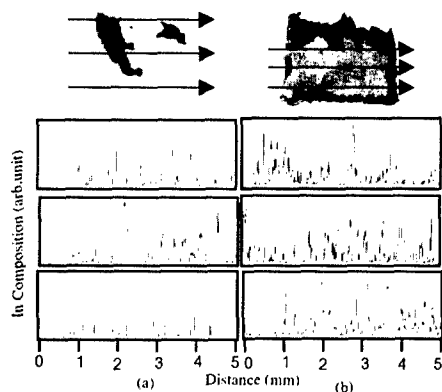


Fig. 5. EPMA line profiles for the needle crystals grown under (a) reduced gravity and (b) normal gravity.

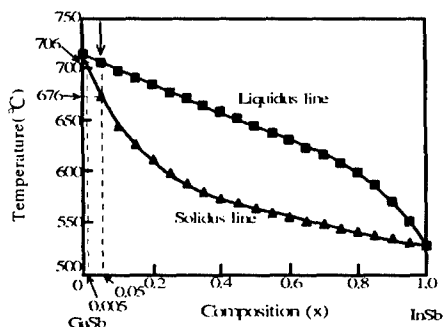


Fig. 6. InSb-GaSb binary phase diagram.

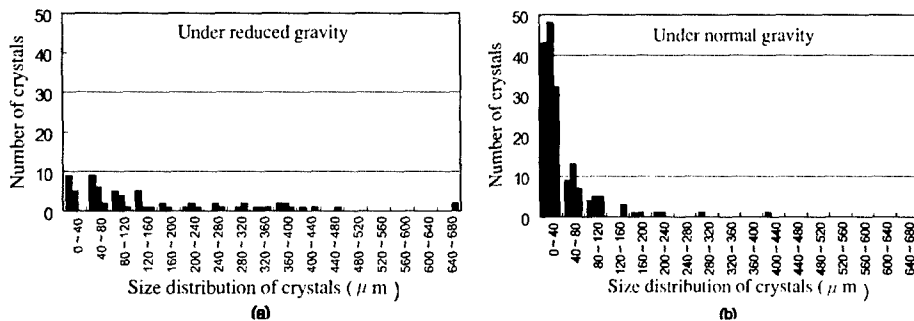


Fig. 7. Size distribution of the needle crystals grown under (a) reduced gravity ( $10^{-3}G$ ) and (b) normal gravity conditions.

### **Formation of projections during InGaSb crystallization**

During the crystallization of InGaSb, many spherical projections were observed on the surface of the sample. The projections emerged out during the crystallization of InGaSb from its melt due to the reason that the density of InGaSb liquid is larger than that of solid. These were similar to the projections observed in the melting and solidification experiment of In/GaSb/Sb done in IML-2 [5]. The projections in this study were found to increase in size slowly. In the present experiment, the drop of the capsule was made to coincide with the appearance and growth of spherical projections so that this process could be observed under microgravity condition. Figure 8 shows the high resolution images recorded using high speed camera during the crystallization of InGaSb in one of the drop experiments. As seen clearly in the images, the spherical projections have just started to appear and grow larger in size slowly as the time passed. The figures (a)-(f) correspond to the images recorded under microgravity condition at the timings 0s, 1s, 2s, 3s, 4s and 4.5 s, respectively. The enlarged image of the final form of the projections formed is shown in the Fig. 9. The projection A was formed under normal gravity condition and the projections B and C were formed

under microgravity condition. The projections formed under microgravity were almost spherical, whereas, the projection formed under normal gravity was not perfectly spherical. Due to gravitational pull, the top surface of the projection A tended to become flat. This shows the influence of gravity on the formation of projections.

It was found that the rate of increase of projection area was high and constant for the first 3 sec. However, in the remaining time, the increase rate became lower and this may be due to the reduction of solute supply in the final stage of projection formation. The solute supply was blocked to some extent by the already formed crystal in the lower portion of the projection.

The In compositions in the formed projection B and C were measured by EPMA and are schematically represented in Fig.10. The In composition in the projection C was found to be more than that of projection B. This is due to fact that the temperature at the time of the formation of projection B was higher than the corresponding temperature of projection C. This indicates that the In composition of the crystallized InGaSb varies depending on the existing temperature at the time of formation. This is in accordance with the InSb-GaSb ternary phase diagram [11]. Since the whole processes of melting and crystallization of InGaSb were recorded using the high speed and high resolution CCD camera (a maximum of 250 images per second) in all the experiments along with the precise values of the corresponding gravity levels and temperatures, it was possible to find the exact conditions at which the projections formed.

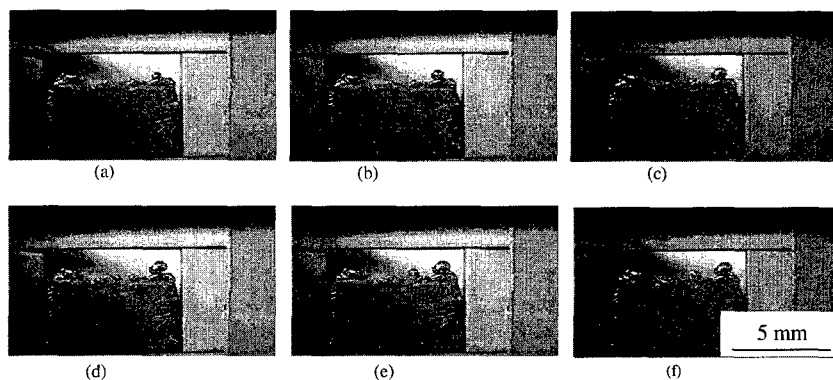


Fig.8. High resolution images recorded during the crystallization of InGaSb at the timings, 0s, (b) 1s, (c) 2s, (d) 3s, (e) 4s and (f) 4.5 s, respectively.



Fig.9. The enlarged images of the final shape of the projections. The projection A was formed under normal gravity condition and the projections B and C formed under microgravity condition.

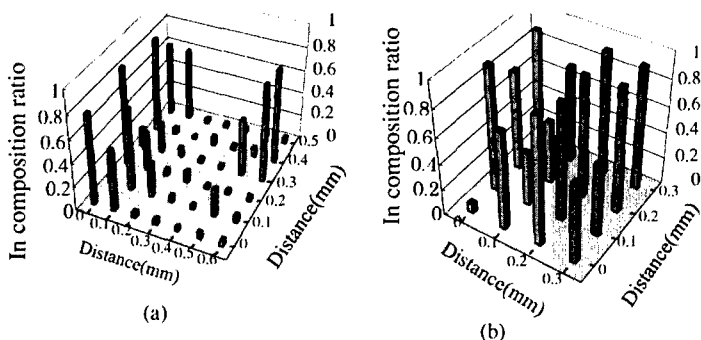


Fig.10.The 3-dimensional schematic representation of the In compositions measured on the (a) projection B and (b) projection C. The measurements were made at the selected points on the squares of areas  $0.6 \times 0.7 \text{ mm}^2$  and  $0.4 \times 0.4 \text{ mm}^2$  of projections B and C, respectively.

## SUMMARY

In the airplane experiment on the crystallization of InGaSb under  $10^{-2}G$  condition, needle crystal was found to have formed and the size of it was larger than the crystal obtained under normal gravity. This showed that the reduced gravity condition is more feasible for crystal growth. In the drop experiment, spherical projections of InGaSb were observed to form during its crystallization under microgravity. On the other hand, projection formed under normal gravity was not perfectly spherical. Indium composition in the projection was found to vary depending on the existing temperature condition during the formation.

## ACKNOWLEDGEMENT

This study is funded by a part of "Ground Research for Space Utilization" promoted by NASDA and Japan Space Forum, and Monbusho grant-in-aid for the International scientific program and scientific encouragement fund in Shizuoka University. Technical assistance of Mr.Momose is acknowledged.

## REFERENCES

1. A.F.Witt, H.C.Gatos, M.Lichtensteiger and C.J.Herman, J.Electrochem.Soc. 125 (1978)1832.
2. A.N.Danilewsky, K.W.Benz and T.Nishinaga, J.Crystal Growth 99(1990)1282.
3. T.Nishinaga, P.Ge, C.Huo, J.He and T.Nakamura, J.Crystal Growth 174 (1997)96.
4. K.Okitsu, Y.Hayakawa, T.Yamaguchi, K.Kumagawa, A.Hirata, M.Tachibana and N.Imaishi, Trans.Mat.Res.Soc.Jpn., 16A (1995) 691.
5. K.Okitsu, Y.Hayakawa, A.Hirata, S.Fujiwara, Y.Okano, N.Imaishi, S.Yoda, T.Oida, T.Yamaguchi and M. Kumagawa, M, Cryst. Res. and Tech., 31(1996) 969.
6. K.Okitsu, Y.Hayakawa, T.Yamaguchi, A.Hirata, S.Fujiwara, Y.Okano, N.Imaishi, S.Yoda, T.Oida and M. Kumagawa, Jpn. J. Appl. Phys. 36(1997) 3613.

7. A.Hirata, K. Okitsu, Y.Hayakawa, Y.Okano, S. Sakai, S. Fujiwara, N. Imaishi, Y. Yamaguchi, S. Yoda, T. Oida and M. Kumagawa, Int. J. Appl. Electromagnetics and Mechanics 10 (1999) 527.
8. A.Hirata, Y.Hayakawa, T.Wada, D.Hirose T.Yamaguchi, Y.Okano, J.Simizu, K.Arafune, N.Imaishi, Y.Kumagiri, X.Zhong, X.Xie, B.Yuan, F.Wu, H.Liu and M.Kumagawa, Proc.of 21st International Symposium on Space Technology and Science (Oomiya, Japan, 1998.5) Vol. II, (1998) 1243.
9. T.Kimura, Y.Hayakawa, T.Ozawa, Y.Okano, A.Hirata, M.Miyazawa, N.Imaishi, K.Arafune, T.Yamaguchi and M.Kumagawa, J.Jpn Soc.Microgravity Appl.15, Supplement II (1999) 472.
10. Y.Hayakawa, Y. Okano, A. Hirata, N. Imaishi, Y. Kumagiri, X. Zhong, X. Xie, B. Yuan, F. Wu, H. Liu, T. Yamaguchi, and M. Kumagawa, J. Crystal Growth 213(2000)40.
11. G.B. Stringfellow, J.Phys.Chem.Solids 33 (1972) 665.